Wet electrostatics and biomolecular self-asembly

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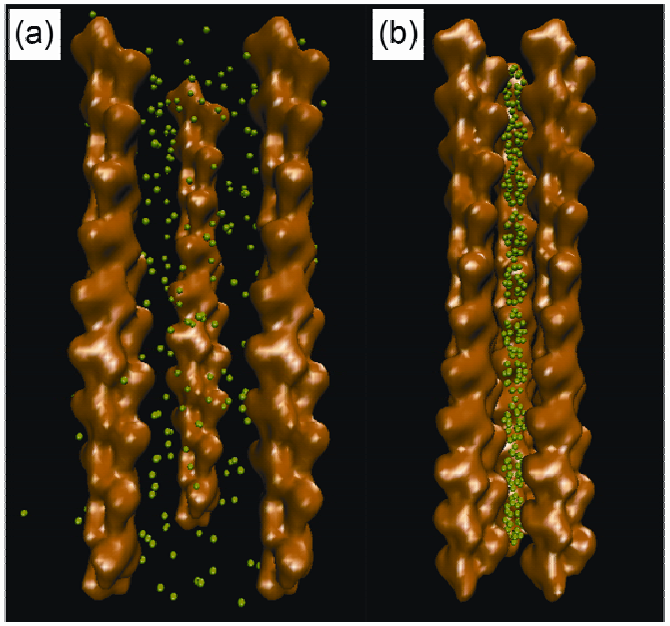
DMR-0409769

Intuitively, two like-charged polymers in water are expected to repel one another. In the presence of multivalent ions, however, such polymers can actually attract one another and form aggregates. The origin of this attraction has been the subject of intense theoretical debate for decades, but no consensus on the mechanism has emerged. Here, we show that ions can mediate attraction between like-charged polymers by condensing into frozen 'ripples' along the length of the polymer. Manipulation of these interactions may lead to therapeutic strategies for cystic fibrosis and a range of other biomedical problems involving aggregation of biopolymers. PNAS **100**, 8634-8637, (2003)



Divalent cations (green) mediate attractions between three anionic F-actin helical polymer rods (gold), by organizing into a frozen ripple of positive charge between the negatively charged rods.

Electrostatics are quite important in biology. All nucleic acids (DNA, RNA) is charged. All cell membranes are charged. Most proteins are charged. While electrostatics are well understood in a vacuum (say for example, Coulomb's Law from High School physics), these interactions are extremely complicated in water. (This is largely due to the long-range nature of the interaction and the entropy of free ions.) This can manifest itself in a number of counterintuitive effects. Intuitively, two like-charged macromolecules in water are expected to repel one another. In the presence of multivalent ions, however, polymers can actually attract one another and form aggregates. The origin of this attraction has been the subject of intense theoretical debate for decades, but no consensus on the mechanism has emerged. Using synchrotron x-ray diffraction, we show that ions can mediate attraction between like-charged polymers by condensing into frozen 'ripples' along the length of the polymer. Manipulation of these interactions may lead to therapeutic strategies for cystic fibrosis and a range of other biomedical problems involving aggregation of polymers. This work was published in PNAS **100**, 8634-8637, (2003).

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Education:

Four undergraduates (James Ho, Brian Heidel, Jessica Bump, and Sam Singh) and three graduate students (Tommy Angelini, Hongjun Liang, John Butler) contributed to this program of research. Sam is still a member of the group. Brian, Jessica, and James have graduated from UIUC, and James is now a graduate student in Bioengineering at U.C. Berkeley. Graduate students Tommy, Hongjun, and John are still in the group working on related experiments. Each has won a number of awards (Fall MRS best poster 2003, UIUC Racheff Award, C&E News *Chemistry Highlight 2003*, APS News *Physics News in 2003*).

Societal Impact:

The ability to manipulate electrostatic interactions in water impinges of a number of outstanding problems. This work may lead to new therapeutic strategies for cystic fibrosis, where negatively charged polymers such as F-actin and DNA bind to and inactivate net positively charged antibiotic proteins, and thereby contribute to long-term infections. An understanding of these electrostatic effects will also potentially lead to improved methods of water purification processes.